LA-DC - 9544 (1968)

PREPRINT FROM

LOS ALAMOS SCIENTIFIC LABORATORY
of the
University of California
LOS ALAMOS • NEW MEXICO

SOILS AND PLANTS AS INDICATORS OF THE EFFECTIVENESS OF A GROSS DECONTAMINATION PROCEDURE

By

E. B. Fowler, J. R. Buchholz, C. W. Christenson, W. H. Adams
Health Division
Los Alamos Scientific Laboratory
Los Alamos, New Mexico, USA

and

Dr. Eduardo Ramos Rodriguez, Jefe
Margarita Celma, Emilio Iranzo, and Dr. Conchita Alvarez Ramis
Division de Medicina y Protection
Junta de Energia Nuclear
Madrid-3, Spain

UNITED STATES
ATOMIC ENERGY COMMISSION
CONTRACT W-7405-ENG. 36

SOILS AND PLANTS AS INDICATORS OF THE EFFECTIVENESS OF A GROSS DECONTAMINATION PROCEDURE

E. B. Fowler, J. R. Buchholz, C. W. Christenson, and W. H. Adams
Health Division
Los Alamos Scientific Laboratory
Los Alamos, New Mexico, USA

and

Dr. Eduardo Ramos Rodriguez, Jefe
Margarita Celma, Emilio Iranzo, and Dr. Conchita Alvarez Ramis
Division de Medicina y Protection
Junta de Energia Nuclear
Madrid-3, Spain

INTRODUCTION

The reports of Dr. E. Ramos and Emilio Iranzo of the Spanish J.E.N. describe the incident in which plutonium was released to the environment.

Five months after the incident, a soils-plant relationship study was initiated to obtain data relative to plutonium concentrations in the soil and the possible resulting concentrations of plutonium in the plants grown on those soils.

The soils are probably composites from several sources and vary from sand to sandy loam in texture; organic matter is generally low. Crops are grown under irrigation and with the addition of fertilizers.

Soils had been removed to various depths in some areas after the incident and replaced by soil known to be free of plutonium from the incident. All areas with the exception of 2-1 had been plowed to a depth of 45 cm. Some areas had been cultivated more than once. This treatment resulted in a complex mixing of the soil profile in the plowed layer.

METHODS

Six sites were selected within the areas known to have been exposed to plutonium; a seventh site (5-3B) about 5 kilometers to the north was selected in an area believed to be free of plutonium. In September 1966 another site (2-3B) about 15 kilometers to the north was sampled; the seventh and eighth sites served as controls. A 50 x 50 meter square served as the sampling site with 9 sampling points on the diagonals. Cores were taken in duplicate at each point at depths of 0-5, 5-15, 15-25, 25-35, and 35-45 cm for a total of 90 samples per site. Vegetation associated with each point was obtained where available.

The complete method of analysis for plutonium will be published elsewhere. In brief, it entailed digestion of an aliquot of the sample (about 50 grams for plant and 1 gram for soils), and centrifugation followed by conversion of the plutonium to the tetravalent state and sorption of the plutonium on AG 1-X2 (Dowex) ion exchange resin (0.15 to 0.30 mm in diameter). The sorbed plutonium was eluted from the column and electroplated for 5 hours. The electroplated plutonium was counted in low background alpha scintillation equipment. The average background of the counters was 0.02 d/m and average recovery from a standard plutonium solution was 75%.

RESULTS

Two observations are important.

(1) In general, the highest concentrations of plutonium were found in the 0-5 cm depth, and

(2) A wide range in plutonium concentrations was found in a single sample from which aliquots were taken.

Average results for each depth and each plot are listed in Table I.

TABLE I. PLUTONIUM CONTENT IN SOILS - AVERAGE (d/m/gram - dry)

Plot No.	Depth (cm) 0-5 Min	0-5 Avg	0-5 Max	5-15 Min	5-15 Avg	5-15 Max	15-25 Min	15-25 Avg	15-25 Max	25-35 Min	25-35 Avg	25-35 Max	35-45 Min	35-45 Avg	35-45 Max	Plot Average All Depths
2-1	0.2	5.6	37	0.2	1.9	8	0.2	6.4	48	0.2	2.6	16	0.2	4.6	46	4.2
2-2	6	294	990	3	80	695	0.2	2.6	9	0.4	2.6	9	0.1	2.4	12	76.7
3-1	0.6	343	4680	0.6	13.8	58	2	27	209	0.4	7.2	53	0.9	18	130	81.8
3-2	1	75	556	1.2	76	268	2	27	198	0.9	2.7	6	0.3	3.3	13	37.0
5-1	0.8	3.1	10	0.7	3.5	14	0.3	3.0	13	0.1	3.3	24	0.2	3.3	11	3.2
5-2	0.1	1.9	9	0.2	1.6	8	0.2	1.2	7	0.1	2.0	11	0.1	3.5	20	2.0
Depth Average All Plots		120			29.5			11.2			3.4			5.9		34.0
Control Plots																
2-3B*	0.3	2.8	8	0.6	1.6	4	ND**	1.0	3	ND**	1.6	2	0.4	2.3	5	1.9
5-3B	0.2	2.0	4	0.1	1.8	7	0.1	7.9	11	0.1	2.8	16	0.1	1.4	5	3.2
Depth Average All Plots		2.4			1.7			4.5			2.2			1.9		2.6

*Samples taken September 1966.
**Not detected

The higher concentration at the 0-5 cm level may result from the manner in which dry, sandy soil breaks over the moldboard of the plow or may be due to later cultivation practices which returned some buried soil to the surface. A study of other methods of burial by plowing is projected.

A study has shown that the wide range in concentration is accounted for by a wide range in particle size. If one assumes a single particle of PuO_2 0.6 μm in diameter, a count of about 0.2 disintegrations per minute results; a particle 18 μm in diameter will produce about 4,680 disintegrations per minute.

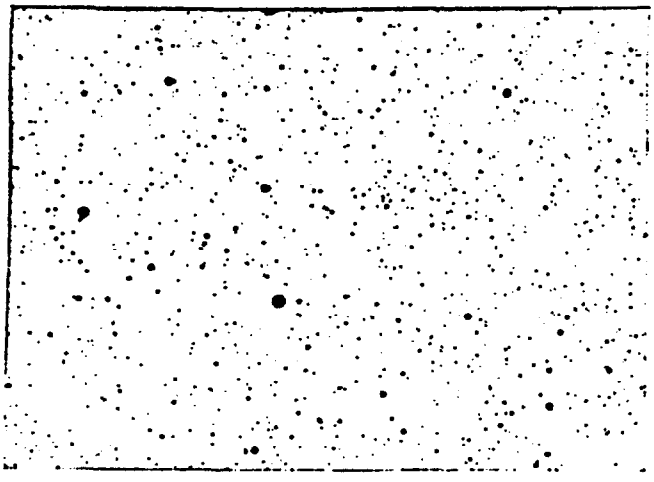

Fig. 1. Particle size range in excavated soils removed from near impact area

Soil which was removed from the site and taken to the USA for burial contained about 10^3 to 10^4 times the amount of plutonium found in the remaining area. The autoradiograph, Fig. 1, shows the observed relative particle size encounted in those soils collected from near the impact area of the device. The range of particle size is obvious and substantiates the observed range in concentrations of plutonium per aliquot of soil sample. Data relative to observed d/m/aliquot from a single sample are presented in Table II. It is believed that these numbers do represent plutonium content in the aliquots, but they also present a problem in attempting to define levels of plutonium content over large areas. A standard approach to the treatment of such data would be of value.

TABLE II. VARIATION IN OBSERVED ACTIVITY IN ALIQUOTS FROM A SINGLE SOIL SAMPLE, PLOT 2-2

Aliquot	1	2	3	4	5	6	7
d/m/gram (dry)	455	778	0	28	8	53	45
Average							195

Cultivated and wild vegetation have been obtained for four different sampling periods from June 1966 through December 1967. Results obtained from 51 samples of tomato fruit and 21 samples of tomato plant are presented in Table III. Data are reported for washed and unwashed plant and fruit obtained from Areas 2-2, 5-1, and 5-2 and from the control area 5-3B. There is some indication that washing the plant by immersion in water removes some activity; however, washing the fruit has little effect on the observed concentration of plutonium.

TABLE III. AVERAGE PLUTONIUM ACTIVITY IN TOMATOES

Treatment	d/m/gram (wet)	Soil Average d/m/gram (dry)
Plots 2-2, 5-1, 5-2		
Washed plant	5.83	27
Unwashed plant	2.82	27
Washed fruit	.0028	27
Unwashed fruit	.0027	27
Plot 5-3B (Control)		
Washed plant	.066	3.2
Unwashed plant	.109	3.2
Washed fruit	.0009	3.2
Unwashed fruit	.0003	3.2
USA Control	.0007	1.2

Factors which might contribute to the observed plant-fruit difference are:

(1) the longer time of exposure of the plant to the environment than the fruit,

(2) the smooth surface of the fruit compared to that of the plant leaf, and

(3) in the case of beans and maize, the natural protection of the bean seed by the pod and maize seed by the husk.

A consideration of the possible health hazard of the average concentration of 0.0028 d/m/gram of wet fruit in comparison to the MPC_w* for plutonium leads to the conclusion that 10^3 kg of tomatoes may be ingested per day without harm; hence a health hazard may be considered nonexistent.

TABLE IV contains data on 24 samples of washed and unwashed maize plants and 13 samples of dry maize seed. A calculation similar to the above would allow for the consumption of 28 kg of maize seed per day without harm. Again, there is no apparent health hazard.

*Maximum permissible concentration in drinking water.

TABLE IV. AVERAGE PLUTONIUM ACTIVITY IN MAIZE

Plots 3-1, 3-2, 5-2

Treatment	d/m/gram (wet)	Soil Average d/m/gram (dry)
Washed plant	.0235	40.3
Unwashed plant	.4234	40.3
Seed (dry)	.1235*	40.3

*Level in 1 sample 12 x average of other 12.

Plot 5-3B (Control)

Washed plant	.0470	3.2
Unwashed plant	.0585	3.2
Seed (dry)	.0863	3.2
USA Control	.018	1.2

Data relative to plutonium concentration in bean plants and seed are reported in Table V. Bean seeds had a higher plutonium concentration than tomatoes by a factor of about 3; however, the health hazard is again nonexistent. Washing removed some surface contamination from the plants. Five samples of seed and 10 samples of plant were analyzed.

TABLE V. AVERAGE PLUTONIUM ACTIVITY IN BEANS

Plots 3-2, 5-2

Treatment	d/m/gram (wet)	Soil Average d/m/gram (dry)
Washed plant	.024	19.5
Unwashed plant	.114	19.5
Seed (dry)	.007	19.5

Results obtained from 20 samples of alfalfa are reported in Table VI. Conclusions are similar to those reported for maize, i.e., a benefit derived from washing and the absence of a health hazard.

TABLE VI. AVERAGE PLUTONIUM ACTIVITY IN ALFALFA

Plot 5-2

Treatment	d/m/gram (wet)	Soil Average d/m/gram (dry)
Washed plant	.0439	2.0
Unwashed plant	.1576	2.0

Plutonium concentrations have been determined on 64 samples of wild plants of a variety of species. Results are presented in Table VII. Plants, such as Gramineae, which produce a seed head of many awns were highest in plutonium concentration. It is probable that the plutonium found is external, since in fruit such as that of prickly pear, the plutonium content of the peel was about 40 times that of the fruit from which the peel had been removed. Lack of rain in the area would allow the particle to remain on the awn for extended periods of time.

TABLE VII. AVERAGE PLUTONIUM ACTIVITY IN WILD PLANTS

Plots 2-2, 2-1

Treatment	d/m/gram (wet)	Soil Average d/m/gram (dry)
Washed plant	27.36	40.9
Unwashed plant	26.63	40.9
Washed fruit	.0037	40.9
Unwashed fruit	.163	40.9
Peeled fruit	.0042	40.9
Peel	.1627	40.9

Plot 2-3B (Control)

Washed plant	.136	1.9
Unwashed plant	.059	1.9

Results of radiochemical analysis for plutonium in water from one well and the associated open storage tank are presented in Table VIII. Water from the wells is used for human consumption as well as for irrigation. The aquifer varies in depth but averages about 50 meters. Plutonium was not detected in water taken directly from the well.

TABLE VIII. PLUTONIUM CONTENT-TANK AND WELL SAMPLES

Sample	Date Sampled	d/m/liter
Well #1		
Direct from Pump	December 1967	Not detected
Tank only	December 1967	Not detected

CONCLUSIONS

Data obtained from the analysis of about 1,300 samples of soil, vegetation, and water for plutonium indicate that no health hazard exists from the intake of such vegetation as tomatoes, maize, alfalfa, or beans grown on the areas where plutonium was released. The absence of a hazard may be credited to the method of decontamination, the initial low levels of plutonium, or both.